INVESTIGATION INTO THE DISCREPANCY BETWEEN MWI AND MWC CDD/CDF EMISSIONS

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INTRODUCTION

On February 11, 1991, the EPA promulgated standards of performance for new municipal waste combustors (MWC's) and emission guidelines for existing MWC's with a unit capacity greater than 250 tons/day of waste. These standards included limitations on total dioxins (tetra- through octa-chlorinated dibenzo-p-dioxins or CDD) and dibenzofurans (tetra- through octa-chlorinated dibenzofurans or CDF). For new units the CDD/CDF stack emission limit was set at 30 nanograms/dry standard cubic meter (ng/dscm) (12 gr/billion dscf), corrected to 7 percent oxygen (dry basis), and was based on use of a spray dryer/fabric filter (SD/FF) emission control system. For existing systems the CDD/CDF emission guideline was established at 125 ng/dscm (50 gr/billion dscf) and was based on use of a dry sorbent injection/fabric filter (DSI/FF) emission control system. In the Federal Register, the EPA concludes that "all types of existing MWC's . . . applying . . . a . . . DSI/FF system can meet a dioxin/furan emission level of . . . 50 gr/billion dscf at 7 percent [oxygen]." Based on limited emissions test data, it was believed that this emission level reflected a nominal 75 percent reduction in CDD/CDF emissions.

The EPA is currently developing emission standards for new and existing medical waste incinerators (MWIs). An initial belief was that MWIs and MWCs equipped with similar air pollution control devices (APCDs) would have similar CDD/CDF emission reductions and stack CDD/CDF removal being effected. This paper compares available CDD/CDF emission data from MWCs and MWIs and examines various parameters which could potentially contribute to higher emissions from MWIs. Based on this examination, a possible explanation was developed involving the partitioning of CDD/CDF between the stack gases and the captured fly ash. Data is then presented from subsequent testing which supports the hypothesis.

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EMISSION COMPARISON: MWI vs. MWC

In the discussion that follows it is important to establish that with similar APCDs, stack CDD/CDF concentrations from MWIs are greater than from MWCs. To accomplish this objective, it is important to institute a frame of reference and a set of terminology reflecting the potential for formation of CDD/CDF in APCDs. Figure 1 illustrates the various chemical processes and the bifurcation of material within the APCD. Both solid and gas phase material exit the furnace (either MWI or MWC) and enter the APCD. CDD and CDF may be in either the gas phase or may be directly associated with solid phase material. Both phases of materials entering the APCD may also provide precursor materials or catalytic surfaces for formation of CDD/CDF in the APCD.

Within the APCD, many complex processes may occur. Surface catalyzed reactions can cause formation of CDD/CDF with key constituents supplied from either the gas phase or from material associated with the particulate or both. When the CDD/CDF, is formed it may be retained on the particle surface or desorbed to the gas phase. Any gas phase CDD/CDF entering the APCD may pass directly through the control device or may be absorbed on solid surfaces. From a mass balance perspective, there is a flow of CDD/CDF into the APCD with additional CDD/CDF formed in the control device. The inflow plus generated CDD/CDF will exit the APCD through the stack or with the collected fly ash. CDD/CDF in the gas phase will exit the APCD with the flue gas while the majority of the solid phase CDD/CDF will exit with the collected fly ash.

Historically, the effectiveness of APCD systems to "control" CDD/CDF emissions has been based on concentration measurements in the stack and at the APCD inlet, ignoring the quantity of CDD/CDF associated with the collected fly ash. The current study examines the available data from MWC and MWI

facilities within the broader framework and attempts to identify process parameters that could be responsible for apparent differences in emission performances between the two classes of incinerators.

APCD INLET CONCENTRATIONS

The initial point of comparison between MWCs and MWIs is to compare the CDD/CDF concentration in the gases leaving the incinerator -entering the APCD system. Figure 2 provides a compilation of inlet CDD/CDF data for a variety of MWIs[1-5] and MWCs[6,7,8]. A relatively wide variation in inlet CDD/CDF concentration is observed indicating differences in equipment design and possibly mode of operation. The important issue, however, is that no trend is observed indicating significantly higher CDD/CDF concentrations coming from either type of combustion equipment.

APCD OUTLET CONCENTRATIONS

There is a relatively small body of data defining the CDD/CDF emission performance of MWIs with APCDs and an even smaller body for units equipped with dry sorbent injection and a fabric filter. One such facility is the MWI at the Borgess Medical Center in Michigan. The Borgess Medical Center incinerator uses dry hydrated lime injection upstream of a baghouse for control of acid gas and particulate matter. A complete description of the Borgess facility and the test program is given in Volume II of the *Michigan Hospital Incinerator Emissions Test Program* [1].

The initial expectation was that CDD/CDF emission rates and APCD collection efficiency for the Borgess facility would be generally consistent with emissions from MWC facilities equipped with DSI/FF. Figure 3 illustrates outlet CDD/CDF concentrations for Borgess[1] and various MWCs[6,7,8] with DSI/FF. The CDD/CDF concentration from all the MWCs tested were under 60 ng/dscm while the outlet concentration measurements at the Borgess MWI ranged between 250 and 650 ng/dscm. Clearly the stack CDD/CDF emission concentrations from Borgess are significantly higher than emissions from MWCs with similar APCDs. Moreover, comparison of the data in Figures 2 and 3 indicates that the "Control Efficiency" of the DSI/FF at Borgess was extremely low and, on certain tests, was negative.

CDD/CDF FORMATION IN APCDs

The above comparisons indicates that stack CDD/CDF emissions from MWIs are higher than from MWCs. Two obvious explanations include the potential that more CDD/CDF is formed in the APCD system of MWIs or that DSI/FF is less effective on MWIs than on MWCs. The following section discusses the possibility that more CDD/CDF is formed in the APCD of medical waste incinerators.

APCD Temperature Formation of CDD/CDF can occur in the APCD and the formation rate generally increases with increasing temperature. Several laboratory studies suggest that peak formation rates occur when the reaction temperature is on the order of 300°C (572°F)[9]. Figure 4 presents the stack outlet CDD/CDF concentration versus APCD temperature for various MWI and MWC facilities utilizing PM control both with dry sorbent injection (DSI) and without acid gas control. The clearly illustrates the trend of increased CDD/CDF emission at higher APCD operating temperature. More importantly, the data tend to fall into two distinct groups. At any given APCD operating temperature, MWIs emit higher CDD/CDF concentrations than MWCs. Based on this comparison the APCD temperature does not provide a reasonable basis for explaining why MWIs have higher CDD/CDF stack emissions.

Surface Area Numerous process parameters have been suggested as key variables influencing low temperature formation of CDD/CDF. Since the basic formation reaction process is believed to be catalytic, one of the key parameters should be the amount of surface area provided by the fly ash. In general, the uncontrolled (inlet to the APCD) particulate matter (PM) loading from an MWC will be about an order of magnitude higher than from a MWI (1-2 gr/dscf for MWCs[10] vs. ~0.1 gr/dscf for MWIs[1,3]). However, the PM emitted by typical MWIs tends to be highly skewed toward submicron particles. Thus, it is possible that the shift in size distribution could more than offset the reduced mass loading.

Only limited size distribution data is available from either MWC or MWI[11] units. By combining actual or typical particle size distridution data with mass loading data it was posible to broadly PM surface area

variation. This process indicated that there is not major difference between MWCs and MWIs as regards the amount of PM surface area available for low temperature formation of CDD/CDF.

Presence of Chlorine and Catalysts The low temperature reactions to form CDD/CDF clearly involve surface reactions, but there is more than one way in which the surface could potentially participate. Researchers have shown a significantly greater formation of chlorinated organics when passing concentrations of Cl_2 rather than HCl over synthetic ash [12]. In these tests, the organic precursor to CDD/CDF was supplied by the particulate, but tests suggest an additional role for the particulate. Specifically, one of the standard processes for forming Cl_2 is to pass HCl over a copper catalyst. Copper or other catalysts in the particulate could enhance formation of Cl_2 and thereby increase CDD/CDF formation. Laboratory experiments using synthetic fly ash have shown that increasing the quantity of copper increased CDD/CDF formation[9]. Further studies examining fly ash from many incinerators found a moderate correlation between copper in ash and CDD/CDF[13].

Based on the above studies, there is at least a possibility that MWIs produce greater quantities of CDD/CDF because of enhanced formation of Cl_2. Medical waste incinerators typically have double the uncontrolled HCl emission compared to MWCs. The difference is due to the higher chlorine content of medical waste. The limited data on the amount of Cu in fly ash is not sufficient to draw conclusions on the comparative role of chlorine and catalysts in the formation of CDD/CDF. Although it remains possible that the higher chlorine content of medical waste (in conjunction with a catalyst) may influence the formation of CDD/CDF, there is a strong opinion that this is not the source of the observed variation in MWI and MWC CDD/CDF emissions. The HCl concentration from MWIs is probably no more than a factor of 2 greater than that from MWCs, and yet the CDD/CDF emissions are increased by a factor of 5 to an order of magnitude.

SYSTEM MASS BALANCE

The preceding evaluation, though not exhaustive, provides no explanation for the observed higher CDD/CDF concentration in MWI stack gases. Those evaluations, however, tend to focus on comparison of inlet and stack outlet CDD/CDF concentrations and do not include consideration of the CDD/CDF associated with the collected fly ash. For a limited number of facilities it is possible to estimate the actual formation of CDD/CDF in APCDs. The calculation requires that a mass balance be performed for the APCD. The amount formed equals the total CDD/CDF leaving the system (both in solid residue and in stack gases) minus the quantity entering the system. While portions of this data are available for many facilities, very few data sets contain all the required data. Three data sets which did contain all the necessary information are Borgess (MWI)[1], Montgomery County (MWC)[6], and Quebec City pilot study[8] (MWC).

For the three facilities with sufficient data, the total CDD/CDF generated is determined by adding all of the exit streams and subtracting the inlet concentrations. Data for all streams were normalized by the volumetric flow rate of flue gas for that facility, corrected to 7% O_2. Results are presented in Figure 5 and show that CDD/CDF formation is consistent between the comparable APCD systems at Borgess (MWI) and Quebec City (MWC) as well as with the DSI/ESP equipped Montgomery County facility. By comparing total CDD/CDF formation in the APCD the broad groupings of data observed in Figure 4 is collapsed into a single line.

GAS/SOLID CDD/CDF SPLIT IN APCDs

The preceding discussion has first shown that there is no significant difference between MWCs and MWIs relative to the concentration of CDD/CDF exiting the incinerator. Next it has been shown that there is no significant difference MWCs and MWIs relative to the mass of CDD/CDF formed in similar APCD systems. Thus, it is strongly suggested that the source of the discrepancy between MWC and MWI stack concentrations is the split between CDD/CDF captured with the fly ash and CDD/CDF which escapes with the flue gas. In general, it is expected that CDD/CDF in the gas phase within the APCD will be released with the flue gas while that associated with the PM (fly ash and sorbent) will likely be captured and exit the APCD with the solid residue. This section examines the issue of CDD/CDF partitioning in MWIs and MWCs.

A common perception is that under the low temperature conditions in an APCD, CDD and CDF will condense onto fly ash or onto injected sorbent material. A brief examination of the CDD/CDF vapor pressure characteristics shows that, in fact, condensation is not the controlling process. Figure 6 illustrates the variation in tetra and octa CDD vapor pressure as a function of temperature[14]. As shown, at 300°F the vapor pressure of octa CDD is > 10^{-3} atmospheres and the vapor pressure of tetra CDD is about 10^{-4} atmospheres. Thus, at 300°F, if the concentration of octa CDD is less than 1000 parts per million, it will remain in the vapor phase or be evaporated if it is a free liquid on the surface of a particle. By comparison, 1000 ng/Nm^3 of tetra CDD is equivalent to a concentration of about 50 parts per trillion.

Based on the above data, it is clear that within APCDs, CDD/CDF does not condense onto the surface of particles and remains as a free liquid or solid. It is also a fact, however, that substantial concentrations of CDD/CDF are found in fly ash collected from MWCs and MWIs. For this to occur it is necessary that the CDD/CDF be chemically or physically bonded to the PM. The implications of this requirement will be examined below.

If CDD/CDF is chemically bound to the surface of particulate matter (chemisorption), one would expect that bonding to be influenced by temperature and by the nature of the particle surfaces. As regards temperature, one would anticipate that the bonds would tend to break as the temperature increases. This possibility is evaluated by examining experimental data from the MWC in Montgomery County, Ohio where CDD/CDF concentrations were determined in both the collected fly ash and stack gas at several different APCD operating temperatures. These data are important in that tests were conducted both with and without sorbent injection. Further, the Montgomery County MWC is equipped with a water quench upstream of the ESP which tends to remove most of the large diameter particulate. In fact, this MWC has a PM size distribution entering the ESP which is quite similar to an MWI controlled-air system.

Table 1 and Figure7 illustrate data from the Montgomery County MWC. The data have been converted such that both the fly ash and stack CDD/CDF concentrations are normalized to the volume of dry flue gas, corrected to 7% oxygen. These data illustrate that greater amounts of CDD/CDF are formed at higher temperatures but also show how temperature impacts the bifurcation. As shown in Figure 7, all of the data without duct injection of sorbent exhibit a linear relationship. As APCD temperature increases, there is a substantial increase in the fraction of the total CDD/CDF which escapes with the stack gas. This is precisely the anticipated trend discussed earlier.Test point TC-5 was the only condition in the Montgomery County test series where hydrated lime was injected into the duct leading to the ESP. Table 1 and Figure 7 illustrate two important trends. First, the total quantity of CDD/CDF leaving the ESP (both gas and solid phase) was significantly reduced relative to the tests without duct sorbent injection at an equivalent temperature (test point TC-4). Additionally, however, of the total CDD/CDF from test TC-5, a much larger fraction was released to the gas phase. In test condition 5, 57% of the total CDD/CDF was released with the stack gas as compared to 22% in test condition 4.

The above described Montgomery County MWC data make two very important suggestions. Reducing APCD temperature will decrease the total quantity of CDD/CDF formed and will also reduce the fraction of that organic which will be released to the gas phase. The other key indication from this single test point is that hydrated lime injection greatly reduces total CDD/CDF formation. Further, the presence of hydrated lime appears to increase the percentage of total CDD/CDF released to the gas phase. The increase in percent released with stack gas was not nearly significant enough to offset the decrease in total formation and, hence, a reduction in stack gas concentration was observed.

Measurements similar to those discussed above for Montgomery County were taken at several other facilities. The Borgess MWI facility tests provide both ash and stack data but the testing covered only a relatively narrow band of fabric filter temperature. For the series of five tests at Borgess, the average CDD/CDF concentration at the APCD inlet, in the stack, and in the ash were 459, 452, and 355 ng/ dscm of flue gas respectively. Thus, the ratio of CDD/CDF in the fly ash to CDD/CDF in the stack gas is 0.78. Since the APCD temperature at Borgess was nominally 320°F, the tendency for CDD/CDF to be retained with the ash is almost identical to that observed at Montgomery County (with duct sorbent injection).

The other facility for which there is a large body of data is the Quebec City MWC which was tested as part of Environment Canada's NITEP program. Table 2 presents the CDD/CDF data in ng/dscm of flue gas basis at several locations in the pilot scale DSI/FF tests. In all cases, there was a substantial concentration of CDD/CDF leaving the fabric filter but essentially all of the organic was retained with the collected

solids. Note that even with the fabric filter operating at 408°F, only 7.3 ng/dscm were in the stack gas as compared to 2383 ng/dscm of total CDD/CDF exiting the fabric filter in the ash and stack gas combined. When compared to the previously described results, the data in Table 2 suggest that perhaps there is something different about the Quebec City Data. Duct injection of sorbent certainly did not significantly suppress the total CDD/CDF formation in the fabric filter. In fact, much more CDD/CDF was formed in the bag house at Quebec City than was formed at Borgess. What is totally different, however, is the fraction of that CDD/CDF that is released to the gas phase.

CARBON LOADING

The current study is unable to prove why the Quebec City MWC retained nearly all of its CDD/CDF with the collected solids. We can, however, suggest that carbon in the fly ash could be a controlling parameter and suggest that this parameter is the key difference between MWCs and MWIs.

Several field tests have demonstrated that injection of small quantities of activated carbon can have a significant impact on the emission of CDD/CDF from both MWC's and MWI's. Activated carbon injection is currently used at a few incinerators for mercury and volatile organic control. Typically, a small amount of carbon is injected into the flue gas and adequately mixed. Effort is made to assure good mixing prior to a moisturizing environment since water is believed to plug the pores and reduce the reactivity of the activated carbon. The small amounts of carbon (typically 20-400 mg/Nm^3 with an average of ~70 mg/Nm^3) are believed to provide a large amount of active surface area for chemisorption of CDD/CDF. Results from a hospital incinerator test in Sweden showed that activated carbon reduced outlet CDD/CDF emissions by 76 to 92% over tests without activated carbon[16]. A full scale MWC in Zurich reduced outlet CDD/CDF by 57 to 93%.

The relevance of the above data to the Quebec City MWC is that the pilot scale tests described in Table 2 were performed prior to completing extensive hardware modifications to improve combustion performance. In fact, personnel from the facility and from Environment Canada described the plume for the Quebec City MWC as containing many "black birds" -- thin, large diameter pieces of black material escaping the ESP. The carbon content of the particulate from this MWC (prior to the facility modification) is not reported in the various Environment Canada (EC) documentation. It is safe to assume, however, that the carbon in the uncontrolled ash was at least at the upper end of the range observed for other MWCs tested in recent years (1 to 5%). EC does report the uncontrolled PM concentration for the pilot scale DSI/FF tests. The average concentration reported was 6700 mg/Nm^3. If the carbon in ash was only 5%, then the total solid phase carbon loading entering the Quebec City pilot-scale fabric filter would be 335 mg/Nm^3. Thus, the "naturally occurring " carbon concentration is, as a minimum, consistent with the level of activated injected into the above MWI in Sweden or the MWC in Zurich.

In contrast to the situation at Quebec City, controlled air incinerators such as the Borgess facility have very low uncontrolled PM concentrations. At Borgess the average carbon content of the fly ash was approximately 5% (not including injected sorbent) and the average PM loading was 253 mg/dscm. Thus, at Borgess the solid phase carbon flow into the fabric filter is only 13 mg/dscm. That is significantly less than at Quebec City. There may be several phenomena which can explain the CDD/CDF retention discrepancy between Borgess and Quebec City, but clearly the flow of solid carbon to the particulate control device is a leading contender. In fact, it is the only phenomena uncovered thus far which can explain the observed discrepancy between MWC and MWI stack CDD/CDF emissions.

TESTING

The Borgess incinerator was retested to evaluate the impact of activated charcoal injection. The system was modified to inject activated carbon upstream of the fabric filter. Eight tests were completed. Three tests were run without carbon injection, two tests with carbon injection at 1 lb/hr, and three tests with carbon injection at 2.5 lb/hr. The test condition averages are illustrated in Figure 8. Carbon injection at 1 lb/hr reduced stack emissions by 88% from baseline average. With carbon injection at 2.5 lb/hr, stack emissions were reduced by 95% from baseline average. The results support the hypothesis that the amount of unburned carbon influences CDD/CDF stack emissions. The complete data set was not yet available to analyze the impacts of carbon injection on the split between captured fly ash and stack gases.

However, it is believed that the observed reduction is due to the carbon adsorbing the CDD/CDF from the gases and transferring it into the captured fly ash stream.

SUMMARY

The preceding material has examined a variety of phenomena in an attempt to explain a major inconsistency between CDD/CDF emissions from MWCs and MWIs utilizing similar pollution control systems and operating under similar conditions. The available data indicates that a larger portion of the CDD/CDF formed in the APCD of MWIs is released with the flue gas. For MWCs the larger fraction appears to be retained with the collected fly ash. The data also indicates that CDD/CDF leaving the APCD with the solid material is chemisorbed to the surface and not merely condensed on the surface. Increasing APCD temperature weakens those bonds and causes more of the CDD/CDF to be released to the gas phase. Further, it was shown that the strength of this bonding appears to depend on the nature of the particulate surface. Injected sorbent material tends to reduce the total quantity of CDD/CDF formed but the sorbent apparently does not provide a strong bonding between the CDD/CDF and the surface.

The issue of surface bonding led to a reexamination of DSI/FF pilot tests at the Quebec City MWC. The data shows three important trends. First, the total amount of CDD/CDF formed in the APCD system at Quebec City is greater than the quantity formed at the Borgess MWI. Secondly, almost all of this CDD/CDF was retained on the particulate matter and not released with the flue gas. This is very different than the situation with MWIs or with the Montgomery County MWC. Finally, it was shown that the Quebec City pilot-tests had a quite high concentration of carbon in the fly ash. In fact, the carbon levels are at least as high as in tests conducted in Europe where activated carbon was injected into the APCD. Those European tests showed major reduction in exhaust CDD/CDF concentration. In contrast to the Quebec City pilot test, typical MWIs (and the MWC test at Montgomery County) have very low solid carbon loading entering the APCD. The carbon in fly ash levels for MWIs are on the same order or less than in MWCs but the total particulate loading in MWIs is about a factor of 10 to 20 less than for MWCs. The low concentration of solid carbonaceous material, with strong bonding to gaseous hydrocarbons like CDD/CDF, could result in more of the formed CDD/CDF being released to the gas phase.

This theory was evaluated by retesting the Borgess incinerator. Tests were conducted with and without activated carbon injection. The results showed that the injection of activated carbon did reduce CDD/CDF stack concentrations to less than 20 ng/dscm @ 7% O_2. Those concentrations are approximately what is expected for a MWC with dry sorbent injection. Therefore, it is believed that the higher CDD/CDF stack emission concentrations observed at MWIs utilizing similar control technologies as a MWC, is due to the lower amount of unburned carbon loading in an MWI. The lower carbon loading in an MWI results in a greater fraction of the CDD/CDF escaping with the flue gas rather than be adsorbed and collected with the fly ash as in an MWC.

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Table 1 Montgomery County CDD/CDF Behavior

* (ng/dscm) corrected to 7% O_2

	Test Condition 1	Test Condition 2	Test Condition 3	Test Condition 4	Test Condition 5	Test Condition 6
Flue gas flow rate (dscm/min)	713	756	922	859	805	779
Oxygen concentration (%)	13.09	13.50	13.61	13.43	12.47	14.50
ESP temperature (°F)	571	396	394	298	306	534
Fly ash collection rate (g/min)	548	398	994	955	2270	436
Fly ash CDD/CDF conc. (ng/g)	2539	1761	2323	1179	9	4399
Fly ash CDD/CDF (ng/min)	1390220	700856	2309592	1125379	20433	1918123
CDD/CDF in fly ash *	3449	1731	4743	2424	42	5304
Stack CDD/CDF emissions *	17109	866	1480	673	57	14517
Total CDD/CDF out *	20558	2596	6223	3097	99	19821
Uncontrolled CDD/CDF conc.*	252	33	38	14	5	214
Total CDD/CDF Generated*	20306	2563	6185	3083	94	19607

Table 2 Complete CDD/CDF Behavior at Quebec City Pilot Study

* (ng/Nm^3) corrected to 7% O_2

Uncontrolled CDD/CDF *	880	2340	2300	1590
Fly ash CDD/CDF*	2076	2589	2576	2376
Stack CDD/CDF*	2.5	0.2	1.1	7.3

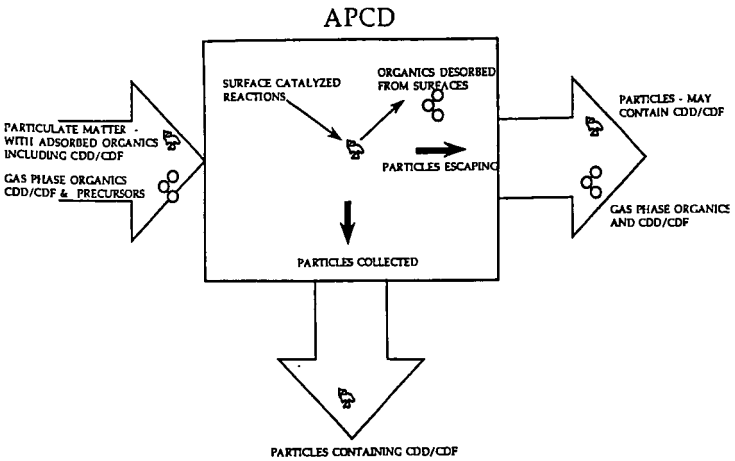


Figure 1 CDD/CDF Behavior in an APCD

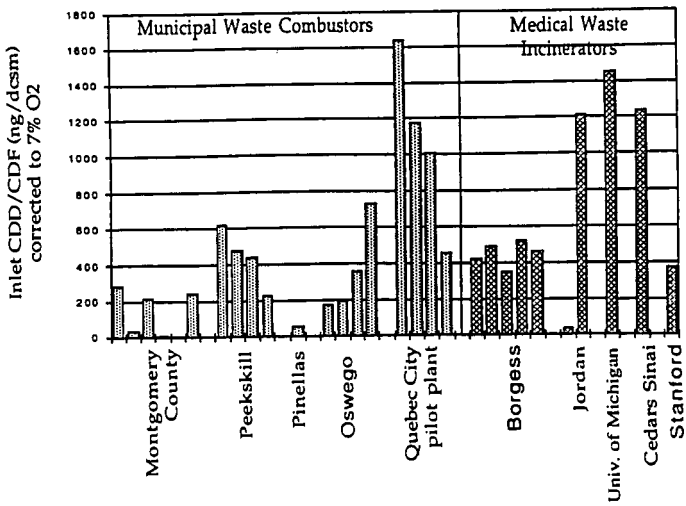
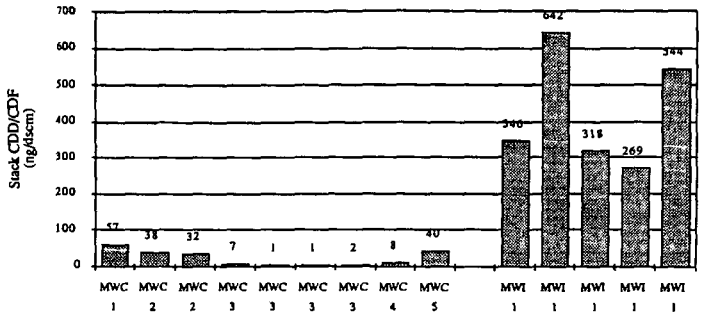


Figure 2 APCD Inlet CDD/CDF Concentrations



Facility code	Facility name	combustor type	APCD
MWC1	Montgomery County, OH	rotary kiln	WQ/DSI/ESP
MWC2	Claremont	mass burn	DSI/FF
MWC3	Quebec City pilot test	mass burn	WQ/DSI/FF
MWC4	St. Croix	MOD/EA	DSI/HE/FF
MWC5	Wurzburg	mass burn	WQ/DSI/FF
MWI1	Borgess	MOD/SA	DSI/FF

Figure 3 Comparison of Outlet CDD/CDF Concentrations for an MWI and MWCs with Dry Sorbent Injection

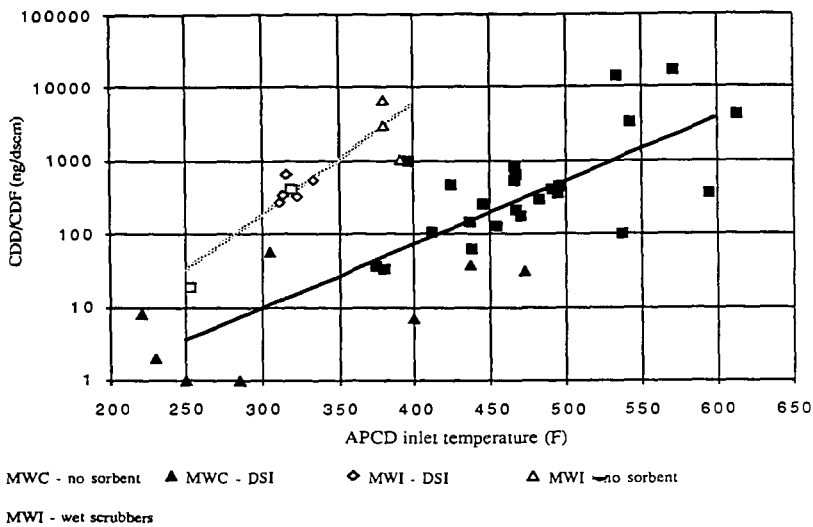


Figure 4 Effect of APCD Temperature on CDD/CDF Outlet Concentration

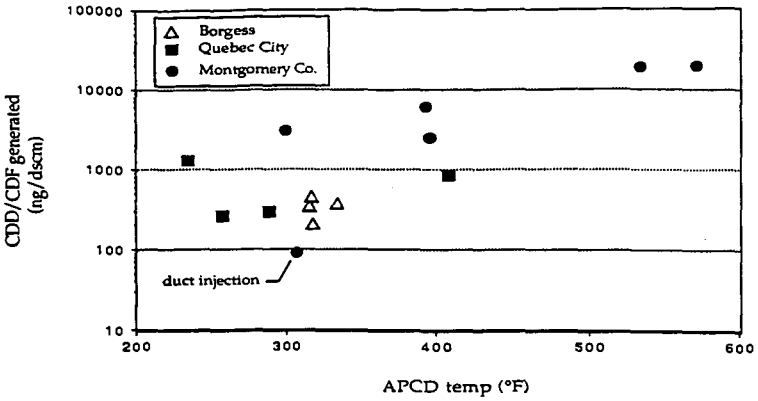


Figure 5 Comparison of Total CDD/CDF Generated in APCDs

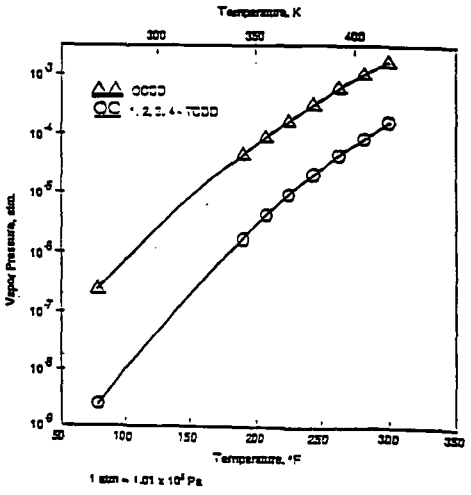


Figure 6 Vapor Pressure of Octa- and Tetra-chlorodibenzo-(p) dioxins

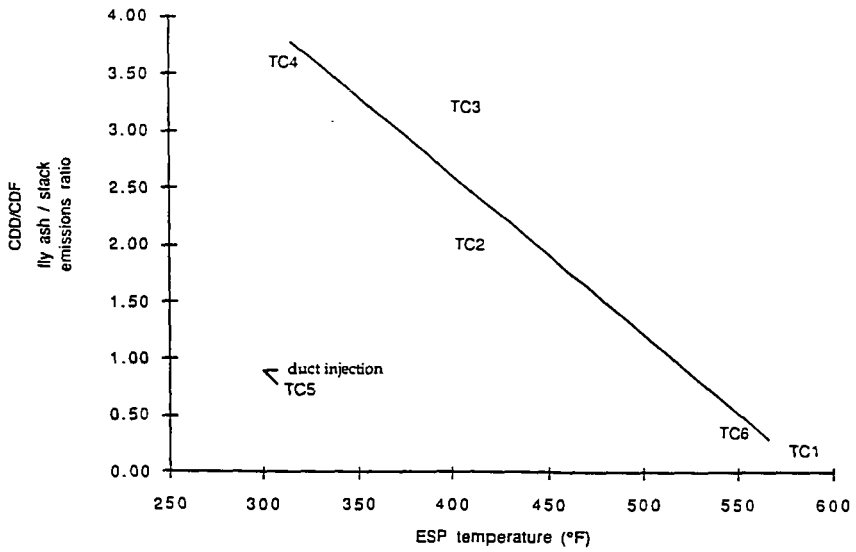


Figure 7 Effect of Temperature on Bifurcation of CDD/CDF in an ESP

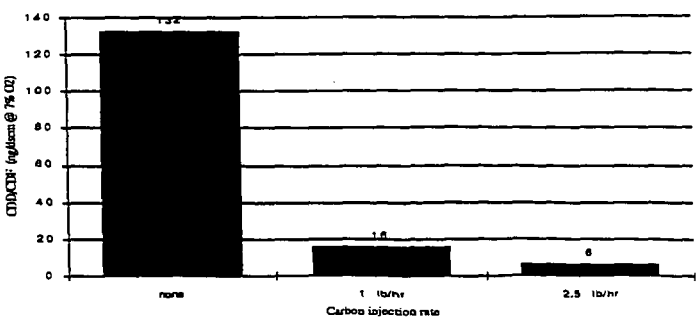


Figure 8 Effect of Activated Carbon on CDD/CDF Stack Concentration